

Feynman variance for neutrons emitted from photo-fission initiated fission chains - a systematic simulation for selected speacal nuclear materials

R. Soltz, E. Hartouni, S. Sheets, A. Glenn

May 21, 2013

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Feynman variance for neutrons emitted from photo-fission initiated fission chains - a systematic simulation for selected special nuclear materials

R.A. Soltz, A. Danagoulian, S. Sheets, A. Glenn, S. Korbly, E.P. Hartouni May 22, 2013

Abstract

Theoretical calculations indicate that the value of the Feynman variance, Y_{2F} for the emitted distribution of neutrons from fissionable exhibits a strong monotonic dependence on a the multiplication, M, of a quantity of special nuclear material. In 2012 we performed a series of measurements at the Passport Inc. facility using a 9-MeV bremsstrahlung CW beam of photons incident on small quantities of uranium with liquid scintillator detectors. For the set of objects studies we observed deviations in the expected monotonic dependence, and these deviations were later confirmed by MCNP simulations. In this report, we modify the theory to account for the contribution from the initial photo-fission and benchmark the new theory with a series of MCNP simulations on DU, LEU, and HEU objects spanning a wide range of masses and multiplication values.

LLNL-TR-637118

Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344.

1 Theoretical Expectation for Y_{2F}

In our Phase I. report we had reported the formula for the Feynman Variance using the induced fission component from [1, 2].

$$Y_{2F} = \left(1 - \frac{1 - e^{-\lambda T}}{\lambda T}\right) \epsilon M_{e} \frac{M_{e} - 1}{\nu - 1} \nu_{2}$$

$$= \left(1 - \frac{1 - e^{-\lambda T}}{\lambda T}\right) \epsilon \left[1 + \frac{(M - 1)(\nu - 1)}{\nu}\right] \frac{\nu_{2}}{\nu} (M - 1)$$

$$(1)$$

where ν_2 refers to the second combinatorial moment for neutrons from a single fission, and ν is the first moment, equal to the mean number of neutrons from a single induced fission. M_e referes to the escape or leakage multiplication, defined as the net gain in neutrons resulting from the fission chain, defined as $M_e = \frac{1-p}{1-p\nu}$, where p is the fission probability, and ν is the mean number of neutrons produced per fission. It differs from the multiplication factor denoted by $M = \frac{1}{1-p\nu}$ in that M_e accounts for the loss of a single neutron for each subsequent induced fission. A useful conversion between the two multiplication factors is given in Eq. 2,

$$\frac{M_e - 1}{\nu - 1} = \frac{M - 1}{\nu} \tag{2}$$

However, Eq. 2 applies to the case of neutron induced fission. For the photon-induced fissions at the Passport facility we need to consider the following additional physics processes:

- 1. the neutron distribution from the initial photo-fission may differ from the neutron-induced fissions that follow in a chain
- 2. the rate of neutrons detected may include a significant contribution from the photodissociation process
- 3. the re-absorption of neutrons by the uranium object and immediate surrounding material

We note that the latter process is not specific to photo-fission initiated chains. As will be explained, this process can be folded into the efficiency term, that is commonly used as a fitting parameter in other applications of neutron time correlations. In this context the variations in the efficiency can impact our measured signal, and therefore must be understood.

Incorporating the different neutron distribution from the initial photo-fission leads to Eq. 6, where ν_{γ} is the mean number of neutrons emitted in a photo-fission, and $\nu_{\gamma 2}$ is the second combinatorial moment.

$$Y_{2F} = \left(1 - \frac{1 - e^{-\lambda T}}{\lambda T}\right) \epsilon \left[1 + \frac{(\nu - 1)}{\nu}(M - 1)\right] \left[\frac{\nu_{\gamma 2}}{\nu_{\gamma}} + \frac{\nu_{2}}{\nu}(M - 1)\right]$$
(3)

The Feynman variance is formed from the ratio of the second moment for the total neutron distribution, Y_2 , to the first Y_1 , which is equal to the mean rate of neutron emission. The photo-neutron dissociation process contributes to Y_1 , but not to Y_2 . Therefore to account for the contribution of dissociation neutrons to Y_{2F} , one needs to multiply by the fraction of neutrons produced from fission events relative to the total.

$$f_d = \frac{\text{neutrons from fission}}{\text{neutrons from fission + neutrons from dissociation}}$$
(4)

Finally, we consider the neutrons that are re-absorbed into the system. In the rate equations developed by Prasad and Snyderman [1], the neutrons that are equivalent to those that leave the object, in that they are no longer available to contribute to subsequent fissions. However, the neutrons that are re-absorbed are not available for detection. This is not a problem for fitting algorithms that fold this into a single overall efficiency factor. For our purposes, the efficiency will need to be known precisely, and therefore we separate this factor, ϵ_l , which we will refer to as the leakage efficiency.

$$\epsilon_l = \frac{\text{fission neutrons emitted}}{\text{fission neutrons emitted} + \text{fission neutrons absorbed}}$$
(5)

We will use ϵ_d for the neutron detection efficiency. The full expression for the Y_{2F} is therefore,

$$Y_{2F} = \left(1 - \frac{1 - e^{-\lambda T}}{\lambda T}\right) \epsilon_d \epsilon_l f_d \left[1 + \frac{(\nu - 1)}{\nu} (M - 1)\right] \left[\frac{\nu_{\gamma 2}}{\nu_{\gamma}} + \frac{\nu_2}{\nu} (M - 1)\right]$$
(6)

The expression for Y_{2F} in Eq. 6 depends on the multiplication as well as the first and second moments for the single fission neutron distributions for neutron induced fissions, ν and ν_2 , and gamma induced fissions, ν_{γ} and $\nu_{\gamma 2}$. The neutron induced moments are measured and parameterized by Zucker and Holden [3]. These are purportedly the numbers that are returned by the MCNP fission package for the default parameter set. For the photo-fission neutron distributions only the first moment (mean) has been published and evaluated. The evaluation is given in the ENDF report Chadwick [4], which references [5] for the data, although the correct reference is [6]. The ENDF evaluation is shown in Fig. 1, along with the corresponding neutron induced fission with the neutron energy shifted by its binding energy.

Because there are no published measurements for the second moment of the photo-fission neutron distribution, $\nu_{\gamma 2}$, we use the MCNP fission package [7] to estimate its value. The photo-fission package for MCNP accepts a neutron distribution parameter, nudist, which can take on values 0–3. We used nudist=3, the default setting, which corresponds to the discrete Zucker and Holden tabulation according to incident neutron energies. However, because the photo-fission model proceeds initially through a (γ, n) reaction, the fission calculation proceeds through the $^{234}\mathrm{U}$ and $^{237}\mathrm{U}$ isotope fissions, which are not tabulated.

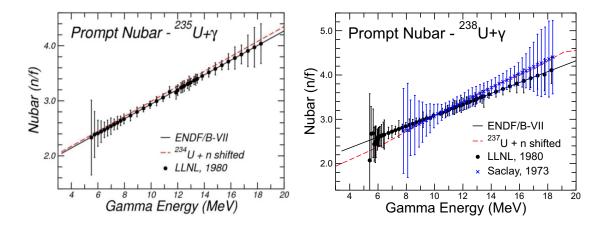


Figure 1: Measurements of mean prompt neutrons from photo-fission, measured by Caldwell et al., and reproduced in an ENDF report by Chadwick et al.

The code defaults to the Terrell approximation [8], for which the mean number of emitted neutrons must be provided. These were obtained from the ENDF database, along with the photo-fission cross-section values used in determining the photon energy distribution for fissions induced by the Passport Beam. The distributions for the Passport beam incident on ²³⁵U and ²³⁸U are shown in Fig. 2. The distribution labeled 0.0 MeV corresponds to the full Passport 9-MeV bremsstrahlung energy distribution, and the mono-energetic distributions are as labeled. This figure also tabulates the mean and second moment neutron distributions calculated two different ways, using a local array and the root histogram package tools.

The measured and calculated values for DU, LEU, and HEU are given in Table 1.

2 Simulation Objects

3 Simulation Details

To test the theory we compare to a set of 36 simulation objects, DU, LEU, and HEU solid spheres spanning 10 grams up to 50 kilograms. The masses, radii, and multiplication values are given in Table 2. The simulations were performed using MCNP version 6.2.24 and DATAPATH variable set to /usr/apps/mcnp/data_lanl-data_data_6.beta2. The spherical SNM object was interrogated with a total of 10^{10} photons distributed randomly in circle transverse to the beam with a radius matching the object in an attempt to approximate a uniform illumination. The emitted neutrons were captured in the wssa format, by specifying an outer cubic rectangle with x and y limits placed at ± 124 cm, and the z limits

Enrichment	$\overline{ u}$	$\overline{\nu_2}$	$\overline{ u_{\gamma}}$	$\overline{\nu_{\gamma 2}}$
DU (0%)	2.43	2.45	2.78	3.09
LEU (20%)	2.45	2.52	2.75	3.04
HEU (93%)	2.51	2.63	2.63	2.90
U-235 (100%)	2.52	2.65	2.62	2.86

Table 1: First and second combinatorial moments for photon and neutron induced fission for various levels of uranium enrichment. Neutron induced moments are calculated from Holden and Zucker tables for 1–2 MeV neutrons reported in the documentation for the mcnp fission package. Photon induced moments are calculated from the photo-fission model with approximate Passport spectrum.

Mass (kg)	radius (cm)	M-DU	M-LEU	M-HEU
0.01	0.500	1.016	1.039	1.064
0.02	0.630	1.020	1.049	1.082
0.05	0.855	1.027	1.067	1.115
0.1	1.077	1.033	1.086	1.149
0.2	1.357	1.041	1.111	1.197
0.5	1.842	1.056	1.155	1.289
1	2.321	1.071	1.202	1.398
2	2.924	1.088	1.265	1.554
5	3.969	1.115	1.390	1.941
10	5.000	1.143	1.532	2.545
20	6.300	1.173	1.754	4.107
50	8.550	1.219	2.298	104.1

Table 2: Mass, radius, and k-factors for du, leu, and heu objects interrogated with 9-MeV bremsstrahlung gamma distribution in simulation.

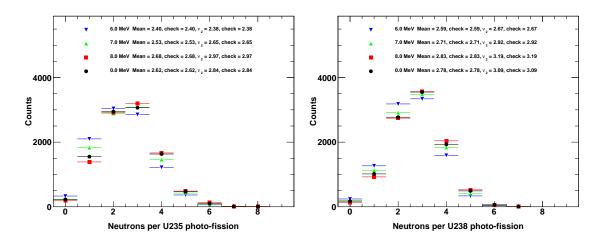


Figure 2: Neutron multiplicity distributions for mono-energetic photo-fission events obtained from fission_v1.8 MCNP package. Distributions are shown for 10,000 photo-fission events for 6, 7, and 8 MeV photons, as well as the full energy distribution of the Passport Beam convoluted with the photo-fission cross-sections. A Poisson distribution with similar mean is also shown for comparison.

placed at [-214,34] cm, thereby approximating an object placed in the top portion of a cargo container vessel. The space around the object is filled with air. The wssa output was converted to text using the read_wssa.x FORTRAN executable that was developed for an earlier project. The Y_{2F} values were calculated for a single time bin of 1 millisecond duration. For this analysis, no energy cuts were applied. For the 50kg HEU object, the wide neutron distribution was rebinned by a factor of 10 to provide a sufficient number of multiple neutron bins to calculate a meaningful value of Y_{2F} .

4 Results

The calculated Y_{2F} values for all 36 objects are plotted in Fig. 3, along with the theoretical curve from Eq. 6, using the values given in Table 1. The other parameters, the fraction of neutrons from fission excluding photo-dissociation, f_d , and the efficiency for neutrons emitted but not absorbed, ϵ_l , are taken directly from the tabulated values reported in the mcnp output files. The theory provides a reasonable description for the DU and LEU objects, but falls beneath the Y_{2F} values for HEU. Because the values for $\nu_{\gamma 2}$ are unmeasured, we also perform a fit to the HEU data in which we allow this parameter to vary. The fitted function is plotted as a dashed red line, and the fitted value of $\nu_{\gamma 2} = 3.81 \pm 0.09$, about 20% higher than the value of 3.09 in Table 1

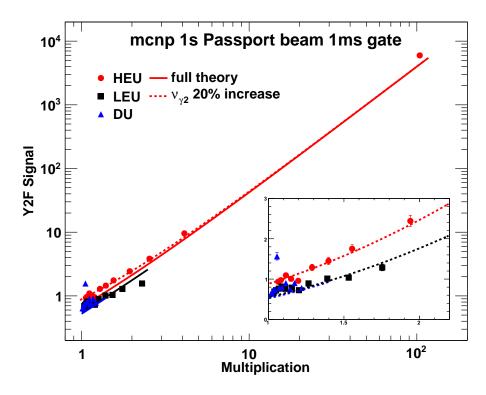


Figure 3: Simulations of DU, LEU, and HEU spherical objects.

5 Conclusions

We have further developed the theory for the Feynman variance for photo-induced fission chains. The revised theory was compared to MCNP calculations for a wide range of fissile objects interrogated by a 9-MeV bremsstrahlung photon beam matching the specifications of the Passport Facility. The theory provides a reasonable description of the simulations results if the second moment of the photo-fission neutron distribution is fit to a value of 3.81 for HEU. This study illustrates the need to obtain reliable and accurate measurements for the neutron distributions from photo-fission reactions for ²³⁵U and ²³⁸U.

References

- [1] M. K. Prasad and N. J. Snyderman, Nucl. Sci. and Eng. 172, 300 (2012).
- [2] S. Walston, LLNL-TR-414245, The Idiot's Guide to the Statistical Theory of Fission Chains.

- [3] M.S. Zucker, N.E. Holden, BNL-38491, 1986, Energy Dependence of Neutron Multiplicity in Fast-Neutron-Induced Fission for 235,238U and 239Pu.
- [4] M. B. Chadwick, et al., Nucl. Instr. Meth. 107, 2931 (2006), ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology.
- [5] J. Caldwell, E. Dowdy, B. Berman, R. Alvarez, and P. Meyer, Phys. Rev. C 21,1215 (1980).
- [6] J. T. Caldwell, R. H. Alvarez, B. L. Berman, E. J. Dowdy, and P. Meyer, Nucl. Sci. Eng. 73,153 (1980).
- [7] J. Verbeke, C. Hagmann, and D. Wright, UCRL-AR-228518, standalone library available from http://nuclear.llnl.gov/simulation.
- [8] J. Terrell, Phys. Rev. 108:783, 1957, Distributions of Fission Neutron Numbers.